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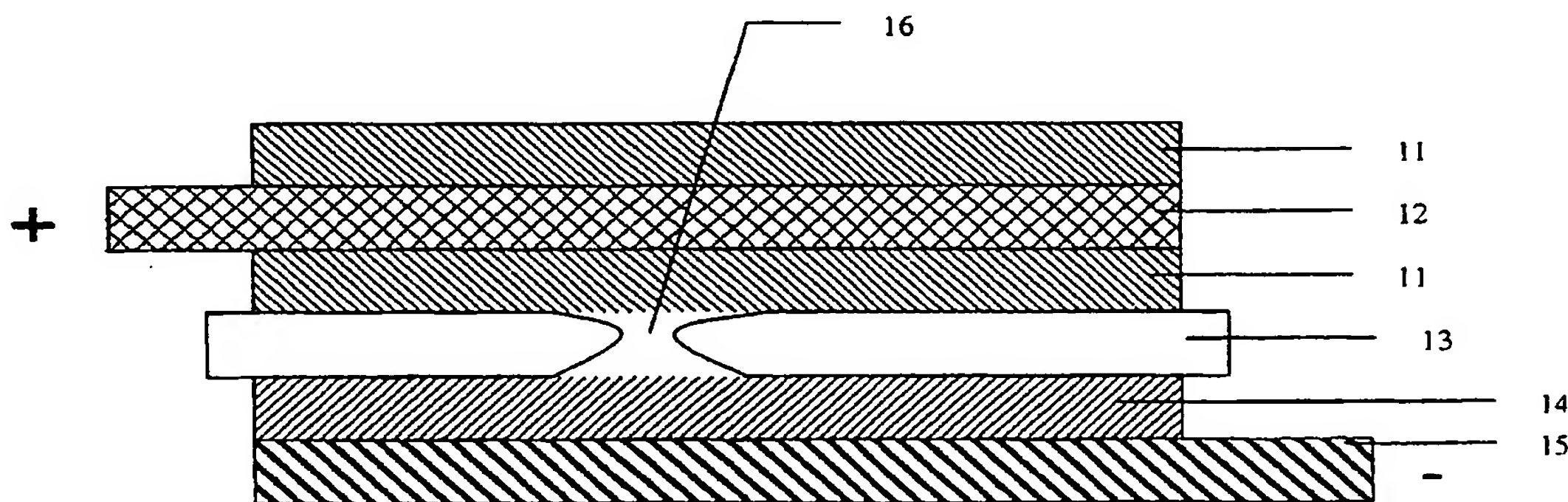
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(54) Title: INFRARED THERMOGRAPHIC METHOD FOR PROCESS MONITORING AND CONTROL OF MULTILAYER CONDUCTIVE COMPOSITIONS



(57) Abstract: An infrared thermographic camera is used to detect non-uniformities in the surface temperature of a multi-layer conductive composition comprising at least two electronically conductive layers (11, 14) separated by an electronically insulating layer (13). The temperature non-uniformities are caused by ohmic heating brought about by a flow of electrical current through a defect (16) or defects in the electronically insulating layer (13) when a predetermined voltage is applied to the two electronically conductive layers (11, 14). The two electronically conductive layers may comprise the positive (11) and negative electrode (14) pair of an electrochemical device, such as a primary or rechargeable battery, supercapacitor, or electrochromic element, and the electronically insulating layer (13), which is superimposed between the two layers (11, 14), may constitute the separator or electrolyte layer of the electrochemical device. One or more layers of the device may additionally comprise electronically insulating liquid components, which may be ionically conductive. The device may be tested "as is", or it may be tested inside a thin, impermeable package. The method is particularly useful for process optimization and control during the fabrication of multi-layer bonded plastic batteries.

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INFRARED THERMOGRAPHIC METHOD FOR PROCESS  
MONITORING AND CONTROL OF MULTILAYER CONDUCTIVE  
COMPOSITIONS

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BACKGROUND OF THE INVENTION

The present invention generally relates to a noninvasive method of testing bonded multi-layer assemblies comprising electronically conductive layers separated by an electronically insulating layer. In particular, the invention includes detecting the spatial location and severity of defects in the electronically insulating layer, where the multi-layer assembly is a bonded preform or a packaged and activated rechargeable plastic lithium-ion battery.

At present, lithium-ion rechargeable batteries are the most advanced widely available energy storage devices. Such devices are typically made of two opposite-polarity, electronically conductive electrodes in the form of long strips or tapes, separated by a continuous strip of a thin microporous, electronically insulating separator. The layers are tightly rolled into a 'jelly-roll' configuration and placed in a metal can canister, which is then filled with an ionically conductive liquid electrolyte solution comprising a mixture of organic solvents and a soluble lithium salt. The electrolyte-activated cell is then sealed and tested.

Li-ion cells have significantly higher volumetric and gravimetric energies than other types of rechargeable batteries, such as lead-acid, nickel-cadmium, or nickel-metal hydride, and have been used commercially with great success since 1991. However, the size, shape and weight of Li-ion cells are strongly influenced by the metal can packaging, which is required to ensure sufficiently high stack pressure to keep the various layers of a Li-ion

cell in close contact. For example, the weight of the metal canister in the case of smaller-capacity cells may approach 30 % of the cell weight. Furthermore, prismatic canisters thinner than about 5-6 mm are very difficult to manufacture, and canisters larger than approximately 50 mm by 40 mm would  
5 have to be made of thicker metal sheet to ensure shape stability, thus increasing the weight of the cell.

The above-mentioned limitations and difficulties were overcome by developing a bonded, or laminated, Li-ion cell technology disclosed in U.S.  
10 Patents 5,456,000, 5,460,904, 5,540,741, and 5,840,087, which are incorporated in their entirety herein by reference. Since the multiple layers comprising a laminated Li-ion cell are strongly bonded to each other, such cells do not require the heavy metal canisters to hold the various layers tightly together and can be packaged in lightweight multi-layer metal-plastic foil  
15 bags. Another advantage of such bonded cells is the fact that such cells can be made in much larger sizes and in any variety of shapes, since there is no limitation on size and shape imposed by the use of a metal canister.

There are several known procedures to fabricate such bonded  
20 electrochemical cells. Bonding of the electrodes to the ionically conductive separator may be achieved by several means, such as by i) casting a solution of a polymer binder and lithium salt on the surface of an electrode followed by evaporation of the volatile casting solvent, ii) casting a solution of a polymer binder, a lithium salt, and a less volatile solvent of the lithium salt on the  
25 surface of an electrode followed by evaporation of the more volatile casting solvent, iii) extruding a melt of a polymer binder with a lithium salt and, optionally, a solvent for the lithium salt directly onto the surface of an electrode, or iv) preparing a self-supporting film of a polymer binder, which may additionally contain a lithium salt, a higher-boiling solvent or other  
30 additives, by casting or extrusion on a neutral substrate, separating the film

from the substrate, applying the separator film to the surface of the electrode and applying heat and pressure to bond the sandwiched layers together.

For example, Feuillade et al. discloses in U.S. Pats. 3,985,574 and  
5 4,086,402 a process for the formation of electrochemical elements in thin layers comprising a polymer-gel separator impregnated with a solvent and saturated with an ionically conductive salt.

Armand et al. describes in U.S. Pat. 4,303,748 a lithium battery, the  
10 electrolyte of which consists of a solid solution of an alkali salt within a plastic, macromolecular, heteroatom-containing material. In U.S. Pat. 4,578,326, Armand et al. teaches novel macromolecular materials constituted by a salt in solution in a copolymer of ethylene oxide and of a second  
15 monomeric unit the structure of which preserves on the chain a polyether structure. The application of the invention is particularly useful in the realization of composite electrodes and electrolyte of primary or secondary electrochemical generators. In U.S. Pat. 5,100,746, Muller et al. describes electrochemical cells containing lithium and a solid polymer electrolyte in the form of thin films, which are produced by making multi-layer assemblies from  
20 double-layer solid polymer electrolyte/composite positive electrode sub-assemblies which are subsequently associated with the other constituent layers of the cell so that the multi-layer assembly so obtained comprises a layer of a metal of low ductility. Cells may be made from multi-layer assemblies by stacking, rolling or bending techniques.

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Lee et al. discloses in U.S. Pat. 4,830,939 a method for forming an interpenetrating polymeric network containing a liquid electrolyte for use in solid state electrochemical cells which comprises forming a mixture of a liquid monomeric or prepolymeric radiation polymerizable compound, a radiation  
30 inert ionically conducting liquid, and an ionizable alkaline metal salt, and

subjecting the mixture to actinic radiation to thereby crosslink the radiation-polymerizable ionically conducting material and form a solid matrix containing the ionically conducting liquid. A similar process is disclosed by Sun in U.S. Pat. 5,603,982, which describes a method for producing solid thin  
5 reinforced terpolymer electrolyte films for use in the fabrication of solid state rechargeable batteries and other solid state electrochemical devices such as supercapacitors, fuel cells, sensors and the like.

Dasgupta et al. describes in U.S. Pat. 5,437,692 a non-aqueous thin  
10 film rechargeable lithium battery using a microporous polymer laminate separator which has been impregnated with an organic liquid electrolyte containing a lithium compound. Both electrodes of the opposite polarity are rendered adherent to the mobile lithium ion carrying electrolyte with a coating of an organic adhesive containing a lithium compound in a concentration  
15 lower than in the electrolyte, disposed between them.

Chern et al. discloses in U.S. Pat. 5,749,927 a continuous process to produce lithium-polymer batteries by extruding the composite electrolyte-electrode sheets onto current collectors to form electrodes. The composite  
20 electrodes, as extruded, are electronically and ionically conductive. The composite electrodes can be overcoated with a solid polymer electrolyte, which acts as a separator upon battery assembly.

In U.S. Pat. 5,811,205 Andrieu et al. describes a bi-functional electrode  
25 for an electrochemical cell or a supercapacitor saturated with a non-aqueous liquid electrolyte, the electrode comprising an electronically conducting porous first layer including at least one first face covered with a microporous second layer constituted by a polymeric material, the electrode being characterized in that the second layer is produced by coagulation of the  
30 polymer from a solution of the polymer impregnating the first face.



An early example of a polymeric electrolyte for electrochromic variable light modulating display devices is disclosed by Reddy in U.S. Pat 4,116,545. The device uses an improved ion conducting layer, which  
5 comprises a polymeric composition containing polyethylene sulfonic acid, a pigment and water, and which is applied as a viscous solution between two flat electrodes of the device.

In U.S. Pat. 3,673,121, Meyer teaches conductive polymer  
10 compositions having a steeply-sloped positive temperature coefficient (PTC) of resistance, which are composed of crystalline polymer having a narrow molecular weight distribution and filled with conductive particles such as carbon black, tin powder, gold powder, silver powder and other conductive particles. An element formed from the compositions is then coated with  
15 electrically conductive coating. Low-resistivity PTC compositions are also disclosed by van Konynenburg et al. in U.S. Pat. 4,237,441. The compositions comprise a crystalline polymer and a particulate filler component which comprises carbon black having a specified particle size and specific surface area. Shaped articles of such compositions are prepared by a process in which  
20 the carbon black is dispersed in the molten polymer, the dispersion is then melt-shaped into thin films, whose two external faces are then adhered to two metal electrodes.

Gozdz et al. disclosed in U.S. Pats. 5,456,000, 5,460,904, 5,540,741,  
25 5,840,087 methods for the fabrication of Li-ion rechargeable battery cell electrode and electrolyte/separator elements formulated as layers of plasticized polymeric matrix compositions, which are laminated to form a unitary battery cell structure. The structure may be stored indefinitely, since it is essentially devoid of electrolyte solution which typically comprises a moisture-sensitive  
30 lithium salt. Prior to the battery's being put into service, at least a portion of

the compatible polymeric composition plasticizer is displaced by contact with a lithium salt electrolyte solution or is removed by extraction with a selective solvent and replaced with the electrolyte solution by simple imbibition.

5           A common feature of the above-described electrochemically active devices is the thin-film, flat configuration of at least two layers of electronically conductive compositions with an interposed thin, bonded layer characterized in that the film is either electronically insulating, or its electronic conductivity is much lower than that of either of the two external layers. The  
10   fabrication of such bonded, multi-layer structures requires that the multiple layers be held together under pressure and/or at elevated temperature for a certain time to affect the bonding, and that many applications require that the electronically insulating layer interposed between the two electronically conducting layers be of as low thickness as possible. It may thus be expected  
15   that material defects, such as trapped air bubbles and larger filler or active material particles, as well as slight variations in the fabrication procedure and the uniformity of temperature and pressure required for effective bonding, would result in direct electrical connection, i.e., short-circuiting, of the external conducting layers through such defects in the middle layer. Such  
20   electrical shorts are typically detected by measuring the DC resistance between the two external layers or, in the case of devices comprising an ionically conductive middle bonding layer, by measuring the AC impedance between the layers.

25           While measurement of electrical resistance provides a fast, convenient, inexpensive, and reliable method of quality control during the fabrication of all the devices listed above, it does not provide any information about the location of the defects. The problem is made more difficult by the fact that in most cases, once bonded, such devices cannot be disassembled into their  
30   constituent layers to facilitate defect location by optical examination. Such



defect location information would be extremely useful in optimizing fabrication procedures and process parameters by indicating, for example, whether the shorting defects are located mostly at the edges of the faulty devices or inside the electrode area. Once the exact location of the defects is indicated, further testing using, for example, microscopic examination of a cross-section of the device at the defect location may reveal its direct cause and lead to changes in process conditions and/or procedures to alleviate the problem.

Therefore, there remains a need in the art for improved methods of detecting defects, particularly the location of defects in insulating layers of multi-layer bonded assemblies so that manufacturing processes may be improved.

## SUMMARY OF THE INVENTION

The present invention provides an improved method of detecting the location of defects in multi-layer bonded assemblies. In particular, the present invention provides an infrared thermographic method for process monitoring and control during manufacturing of multi-layer conductive compositions, in particular, of multi-layer, flat-plate electrochemical energy-storage and electrochromic devices. More specifically, the present invention provides a sensitive, accurate, fast, reliable, real-time, and automated method for detecting and providing precise location of low-electrical-resistance defects in bonded multi-layer polymer- or plasticized-electrolyte batteries, supercapacitors, and electrochromic windows at various stages of their assembly, packaging and testing.

The present invention relies on the fact that the internal layer of the devices is electronically insulating and is interposed between two layers of

electronically conducting sheet material. Therefore, when an alternating or unipolar electrical voltage is applied to the two electronically conductive external layers and a defect provides an electronically conductive path between the conducting layers, the electrical current will flow exclusively through the very small electrical defect area. The small defect area will cause a relatively high electrical current to flow through the defect, causing a local heating effect according to the well-known formula for the amount of heat,  $W$ , dissipated by an electrical current flowing through a resistive element:  $W = U \times i^2$ .

The method of the present invention makes it possible for the first time to localize unequivocally microscopic defects in large-area multi-layer bonded structures and thus to introduce well-informed changes and modifications to the manufacturing equipment and operating procedures to minimize the occurrence of such defects, thereby to increase the yield of the desired product.

Additional objects and advantages of the invention will be set forth in part in the detailed description which follows, and in part will be obvious from the description, or may be learned by practice of the invention. The objects and advantages of the invention will be obtained by means of instrumentalities in combinations particularly pointed out in the appended claims.

#### BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a perspective view of two multi-layer bonded structures which may be tested in accordance with the method of the present invention.

FIG. 2 is a schematic cross-sectional view through an electrical short-circuit point.

FIG. 3 is a thermographic image of the location of two point defects in a multi-layer unextracted plastic Li-ion battery laminate.

5                    DETAILED DESCRIPTION OF THE INVENTION

One of the unique characteristics of solid-polymer- and plasticized-polymer-electrolyte primary and rechargeable lithium cells, whether based on metallic lithium, lithium alloy, carbon, or other intercalation negative  
10 electrodes is that they can be produced in a very thin film form, i.e., about 100  $\mu\text{m}$  and up (see for example, U. S. Pat. 4,517,265). All such electrochemical energy storage devices, as well as some supercapacitor structures and electrochromic mirrors and windows, have an essentially similar construction comprising of at least two electronically conducting thin-sheet electrodes  
15 intimately attached or bonded to two opposite surfaces of an interposed non-electronically conducting thin-sheet separator material which may, or may not, be in an ionically conducting state, which can be attained through the inclusion of i) a molecularly dispersed solid salt of lithium or ii) of a solution of lithium salt in appropriate solvents.

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For the purpose of insuring that the component layers of the devices stay in a proximate contact even when external pressure is not applied during use, fabrication of the devices often involves passing the separate layers through one or more pairs of rollers which may or may not be heated to effect  
25 the permanent bonding between the layers. In another, often used process, a multi-layer stack of separate component layers of the device is placed between two platens of a hydraulic or mechanical press and are held together for a predetermined length of time, often with a simultaneous application of heat, to effect the bonding. Under such high pressure and temperature conditions, the  
30 component materials of the various layers become soft and undergo

compression and lateral flow which, while advantageous to cause the desired degree of interlayer bonding, may cause local excessive flow of the layers having a lower viscosity and may thus result in an excessive local thinning of such layers or even in the direct contact of the two outside, higher viscosity layers. Such direct contact of the two outside electronically conductive layers through the interposed electronically insulating layer is highly undesirable, since it causes the device to become non-functional.

One example of such a multi-layer electrochemical device is the plastic lithium-ion rechargeable cell disclosed in U. S. Pat. 5,840,087; as shown further in FIGS. 1a and 1b. The simplest variant of the devices is fabricated by first laminating an aluminum grid current collector, which was previously treated by a process disclosed in U. S. Pat. 5,554,459 onto a positive-electrode sheet, such as that described in U.S. Pat. 5,456,000. In parallel, a counter-electrode is fabricated by laminating a copper current collector grid treated by a process disclosed in U. S. Pat. 5,840,371 onto a negative-electrode sheet such as that described in U.S. Pat. 5,456,000. In the final assembly step, a plasticized-polymer separator film, such as that described in U. S. Pat. 5,418,091, is interposed between the two the current collector-electrode subassemblies and the three-layer stack is passed through a heated double-roll laminator or placed between two metal platens on a heated press for final bonding. More complex multi-layer assemblies, similar to the one described above, are fabricated by an analogous process described in more detail in the above-listed U. S. patents.

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In particular, Fig. 1a shows a positive-electrode sheet comprised of two positive electrode layers 11, laminated together with a positive current collector grid 12. The negative-electrode sheet comprises a negative electrode layer 14, laminated to negative current collector 15. The positive-electrode

sheet and negative-electrode sheet are then bonded together with separator film 13, providing electrical insulation.

Fig. 1b shows a three layer structure comprised of two positive-electrode sheets, each made up of two positive electrode layers 21, laminated together with a positive current collector grid 22, and a negative-electrode sheet, made up of two negative electrode layers 24, laminated to a negative current collector 25. The positive-electrode sheets and negative-electrode sheet are bonded together with separator films 23, providing electrical insulation. Alternatively, a similar structure having two negative-electrode sheets and one positive-electrode sheet could be utilized.

Effective bonding between the layers is possible only when both bonded surfaces are heated to a temperature close to their melting or softening point and, simultaneously pressed together. It is thus often observed in practice that electrical short-circuit defects were created under such temperature and pressure conditions in areas where the thickness of the interposed, electronically insulating layer is insufficient, or if the layer contained entrapped air bubbles, or if one of the external electrode subassemblies contained a protruding, conductive particle. In addition, such defects can be created when a multi-layer assembly comprising a number of individual sheets whose leading edges are slightly off-set, are fed into a narrow gap of a double-roller laminator equipment. The leading edge of the off-set layer or layers will be hard-pressed into the extended electronically non-conducting layer, thus increasing the probability of a short-circuit through the layer at the edge line. Irrespective of their underlying cause, all short-circuited elements are non-functional and must be discarded, thus decreasing the yield of the desired product and increasing its cost.

Fig. 2 is a schematic cross-sectional view of an electrical short-circuit point in a multi-layer bonded structure as shown in Fig. 1a. In particular, Fig. 2 shows a defect 16, in the separator film 13, which results in electrical contact between the positive-electrode sheet and the negative-electrical sheet, or in other words a short-circuit.

While short-circuited preforms at various stages of fabrication of laminated cells can readily be tested by measuring electrical resistance between two electronically conducting layers, such measurements do not provide any information about the location of the defect. It should be apparent that such information would be very useful in identifying the underlying cause or causes of the defects and in devising countermeasures to prevent their formation. The present invention provides an effective and practical solution to this problem.

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It has been discovered that even a high-resistance, so-called 'soft short', point defect between the two conductive layers with an ohmic resistance of 100-200 kohm generates sufficient local increase in temperature of the surrounding material to become clearly detectable with a commercial-grade thermography camera, such as Avio TVS-2000 manufactured by Nippon Avionics, Ltd., which has a nominal temperature resolution of 0.1°C, when a DC voltage of 10-20 V is applied to the conducting layers. This implies that as little as 1-2 mW of dissipated power can be detected in such multi-layer structures by causing an increase of about 0.2-0.3°C in local temperature.

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When a sample with a lower-resistance electrical short-circuit defect was studied, an applied voltage of less than 0.5 volt was sufficient to visualize the location of the defect. It was also discovered that the very low current (of the order of several mA) flowing through such a detectable defect is stable in time and does not cause further degradation of the device. Of course, if a less

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sensitive camera were used, the current might have to be increased to the level where the local heating effect would exceed thermal stability of the component materials and result in their thermal degradation.

5           It was further discovered that point-like and line-like defects were equally easy to detect and localize in multi-layer laminated plastic battery structures comprising an unextracted plasticizer, such as those described in U.S. Pats. 5,456,000, 5,460,904, 5,540,741, and 5,840,087, which are incorporated in their entirety herein by reference. Additionally, electrical  
10 short-circuit defects were also readily detected and localized in finished flat-plate plastic Li-ion batteries comprising multi-layer structures composed of up to 13 component layers saturated with a solution of a lithium salt in an appropriate liquid solvent and packaged in multi-layer metalized-plastic bags. While such electrically short-circuited cells were not capable of retaining a  
15 constant voltage after the initial charge, attempted charging at a customary rate of charge of 0.5-1C, which is equivalent to a current density of 1-3 mA/cm<sup>2</sup>, caused a sufficiently high shorting current to flow through the defect area to cause a temperature increase of the external packaging material that was readily detectable with a thermographic camera.

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Illustrative examples of several configurations of multi-layer bonded structures, which are well suited for testing in accordance with the method of the present invention and the results of such testing, are as follows:

25   Example 1

A separator/electrolyte membrane coating solution was prepared by suspending 30 parts by weight of an 88:12 vinylidene fluoride (VdF):hexafluoropropylene (HFP) copolymer of about  $380 \times 10^3$  MW (Kynar  
30 FLEX 2801, Elf Atochem) and 20 parts silanized fumed silica in about 200

parts acetone and adding to this mixture about 40 parts dibutyl phthalate (DBP) plasticizer. The completed mixture was warmed to about 50°C to facilitate dissolution of the copolymer and was homogenized in a laboratory ball mill for about 6 h. A portion of the resulting slurry was coated on a glass plate with a doctor blade device gapped at about 0.3 mm. The acetone coating vehicle was allowed to evaporate within the coating enclosure under moderately flowing dry air at room temperature for about 10 min to yield a tough, flexible, plasticized film which was stripped from the glass plate. The film was about 0.1 mm thick and was easily cut into rectangular separator elements.

A positive electrode composition was prepared by homogenizing in a lid-covered stainless steel blender for about 10 min at 2500 rpm a mixture of 65 parts by weight of 53- $\mu$ m sieved  $\text{Li}_{1.05}\text{Mn}_{1.95}\text{O}_4$  prepared in a manner described in U.S. Pat. No. 5,266,299, 10 parts VdF:HFP copolymer (Kynar FLEX<sup>®</sup> 2801, Elf Atochem), 18.5 parts dibutyl phthalate, 6.5 parts conductive carbon (Super-P Black, MMM Carbon, Belgium), and about 100 parts acetone. The resulting slurry was degassed by briefly applying a reduced pressure to the mixing vessel, and a portion was then coated on a glass plate with a doctor blade device gapped at about 0.4 mm. The coated layer was allowed to dry within the coating enclosure under moderately flowing dry air at room temperature for about 10 min to yield a flexible, plasticized film which was stripped from the glass plate. The film, comprising about 65% by weight of particulate active intercalation material, was about 0.12 mm thick and was easily cut into rectangular electrode elements.

A negative electrode composition was prepared by homogenizing in a lid-covered stainless steel blender for about 10 min at 2500 rpm a mixture of 65 parts by weight commercial petroleum coke (MCMB 25-10, Osaka Gas Co.), 10 parts VdF:HFP copolymer (Kynar FLEX<sup>®</sup> 2801, Elf Atochem), 21.75

parts dibutyl phthalate, 3.25 parts Super-P conductive carbon, and about 100 parts acetone. The resulting slurry was degassed, and a portion was then coated on a glass plate with a doctor blade device gapped at about 0.5 mm. The coated layer was allowed to dry within the coating enclosure under  
5 moderately flowing dry air at room temperature for about 10 min to yield a tough, flexible film which was readily stripped from the glass plate. The film, comprising about 65% by weight of particulate active intercalation material, was about 0.15 mm thick and was easily cut into rectangular electrode elements.

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### Example 2

Bonded, unextracted battery cell laminates depicted in FIGS. 1a and 1b were fabricated as described in detail in U.S. Pat. 5,840,087. After the final  
15 lamination step, the electrical resistance of the multi-layer laminated structures was tested at 1 kHz and recorded. Samples having the structure shown in FIG. 1a which exhibited electrical resistance less than about 20 kohm, and samples having the structure shown in FIG. 1 b which exhibited electrical resistance less than about 10 kohm were each divided into three groups.

20

A first of these groups of two types of cells was left for further testing. The second group of two types of cells was subjected to exhaustive extraction of the DBP plasticizer as disclosed in U.S. Pat. 5,540,741, dried in air at 60°C for 1 h, and subjected again to electrical resistance measurement at 1 kHz.

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The third group of two types of cells was packaged in impermeable metalized plastic laminate bags with two protruding electrical tabs, then activated with a liquid-electrolyte solution containing  $\text{LiPF}_6$  at a concentration of 1 M, sealed, degassed under vacuum, and subjected to an initial charging  
30 cycle at a C/2 rate with an upper cutoff voltage of 4.5 V. Cells which showed

signs of internal shorts by exhibiting longer than expected charging times to reach a predetermined capacity were separated for testing.

### Example 3

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A multi-layer unextracted plastic Li-ion cell laminate having the structure as depicted in FIG. 1a and exhibiting DC resistance of 1.2 kohm was placed on a laboratory bench in the field of view of a thermographic camera Avio TVS-2000. The two metal tabs of the battery preform were connected to a regulated power supply (HP E3610A) set to a maximum current of 0.5 A. As shown in the thermographic image of Fig. 3, when the voltage applied to the terminals of the battery preform was slowly raised to 1.9 V, two small areas located at the edges of the preform appeared on the screen, indicating a slightly raised temperature (no more than 0.2-0.3°C above the environment's temperature) at these two locations, and indicating the exact location of the two electrical short-circuits. Microscopic examination indicated that in one case, the electrical short was caused by a wire-like non-flattened protrusion left after cutting through the metal grid. In the second case, the battery preform was trimmed too closely through all the layers, with some grains of the electronically conducting materials connecting the two opposing electrodes.

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### Example 4

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A multi-layer extracted and air-dried plastic Li-ion cell laminate having the structure as depicted in FIG. 1b and exhibiting DC resistance of 120 kohm was placed on a laboratory bench in the field of view of a thermographic camera Avio TVS-2000. The common negative (copper grid) tab and one of the two positive (aluminum grid) metal tabs of the battery preform were connected to a regulated power supply (HP E3610A) set to a

30

maximum current of 0.5 A. When the voltage applied to the terminals of the battery preform was slowly raised to about 15 V, one small brighter area located in the middle of the external positive electrode appeared on the screen, indicating a slightly raised temperature (no more than 0.2-0.3°C above the environment's temperature) at this location, indicating the exact location of the electrical short-circuit. Microscopic examination revealed the presence of a very short section of an aluminum wire derived from the grid trimming operation and embedded in the top-most positive electrode.

#### 10 Example 5

A multi-layer extracted and air-dried plastic Li-ion cell laminate having the structure as depicted in FIG. 1b, packaged, activated with a liquid electrolyte solution, and tested as described in Example 1 was found not to hold charge voltage over an extended period of time (days). The cell exhibited AC resistance of 0.15 ohm at 35 kHz (at  $Z'' = 0$ ). The packaged cell was placed on a laboratory bench in the field of view of a thermographic camera Avio TVS-2000. The common negative (copper grid) tab and the two positive (aluminum grid) metal tabs of the cell were connected to a regulated power supply (HP E3610A) set to a maximum current of 0.2 A. When the cell voltage applied to the terminals of the cell was slowly raised to about 2.5 V, one small brighter area located at the edge of the cell packaged inside the bag and a brighter line connecting the two terminal tabs of the battery through the multi-layer packaging foil appeared on the screen. The single point indicated the location of an edge defect in the battery preform, and the line defect was found to be caused by an electrical short-circuit between the cell terminals through the aluminum layer of the packaging foil caused by too high a sealing temperature in the cell packaging step.

Example 6

A multi-layer extracted and air-dried plastic Li-ion cell laminate having the structure as depicted in FIG. 1b, packaged, activated with a liquid electrolyte solution, and fully charged as described in Example 1 was placed on a laboratory bench in the field of view of a thermographic camera Avio TVS-2000. When the common negative (copper grid) tab and the two positive (aluminum grid) metal tabs of the cell were electrically short-circuited for 1-2 seconds with a thick copper wire, many areas of the cell, especially along one long side of the preform, and in the metal tab area, appeared warmer than average. Further examination of the causes of the non-uniformity of heating was an uneven pressure distribution applied by the rollers of the laminating equipment to the various components of the cell during the final lamination step. This uneven pressure caused an excessive thinning of the plasticized polymer separator on one side of the preform, which resulted in its higher ionic conductivity and higher shorting current density in this area.

Comparative Example 1

A multi-layer unextracted plastic Li-ion cell laminate having the structure as depicted in FIG. 1a and exhibiting a DC resistance of  $>8$  Mohm at 1 kHz was placed on a laboratory bench in the field of view of a thermographic camera Avio TVS-2000. The (copper grid) tab and the positive (aluminum grid) metal tabs of the battery preform were connected to a regulated power supply (HP E3610A) set to a maximum current of 0.5 A. Even when the voltage applied to the terminals of the battery preform was slowly raised to about 20 V, no surface temperature non-uniformities were observed, indicating an absence of electrical defects in the preform.



Comparative Example 2

A multi-layer extracted and air-dried plastic Li-ion cell laminate having the structure as depicted in FIG. 1b and exhibiting DC resistance of 6.5 Mohm was placed on a laboratory bench in the field of view of a thermographic camera Avio TVS-2000. The common negative (copper grid) tab and one of the two positive (aluminum grid) metal tabs of the battery preform were connected to a regulated power supply (HP E3610A) set to a maximum current of 0.5 A. Even when the voltage applied to the terminals of the battery preform was slowly raised to about 20 V, no surface temperature non-uniformities were observed, indicating an absence of electrical defects in the preform.

It is anticipated that other embodiments and variations of the present invention will become readily apparent to the skilled artisan in the light of the foregoing specification. Such embodiments and variations are intended to likewise be included within the scope of the invention as set out in the appended claims.

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What is claimed is:

1. A method of testing, said method comprising the steps of:  
providing a device to be tested, said device comprising at least two electronically conductive layers separated by an electronically insulating layer;  
providing an infrared thermographic camera;  
placing said device in the field of view of said thermographic camera;  
applying a predetermined current and voltage to said conductive layers of said device;  
observing through said thermographic camera any surface temperature non-uniformities in said device.
2. A method according to claim 1, wherein said device comprises a multilayer electronic device.
3. A method according to claim 2, wherein said electronic device comprises a multilayer electrochemical device.
4. A method according to claim 3, wherein said device comprises a positive electrode sheet and a negative electrode sheet with an insulating separator layer therebetween.
5. A method according to claim 3, wherein said device comprises a first positive electrode sheet, a negative electrode sheet, a first insulating separator layer between said first positive electrode sheet and said negative electrode sheet, a second positive electrode sheet, and a second insulating separator layer between said negative electrode sheet and said second positive electrode sheet.

6. A method according to claim 3, wherein said device comprises a first negative electrode sheet, a positive electrode sheet, a first insulating separator layer between said first negative electrode sheet and said positive electrode sheet, a second negative electrode sheet, and a second insulating separator layer between said positive electrode sheet and said second negative electrode sheet.
7. A method according to claim 2, wherein said device is selected from the group consisting of primary batteries, rechargeable batteries, supercapacitors and electrochromic elements.
8. A method according to claim 7, wherein said device is a multi-layer bonded rechargeable plastic lithium ion battery.
9. A method according to claim 1, wherein said predetermined current is sufficiently low that no degradation of said device is caused.
10. A method according to claim 9, wherein said predetermined current is about 0.5 A.
11. A method according to claim 1, wherein non-uniformities are increases in temperature surrounding a defect area in said insulating layer.
12. A method according to claim 1, wherein said device is tested in the absence of any other materials.
13. A method according to claim 1, wherein said device is packaged inside a thin, impermeable package prior to testing.

14. A method of determining the location of short circuits in a device comprising at least two electronically conductive layers separated by an electronically insulating layer, said method comprising the steps of:

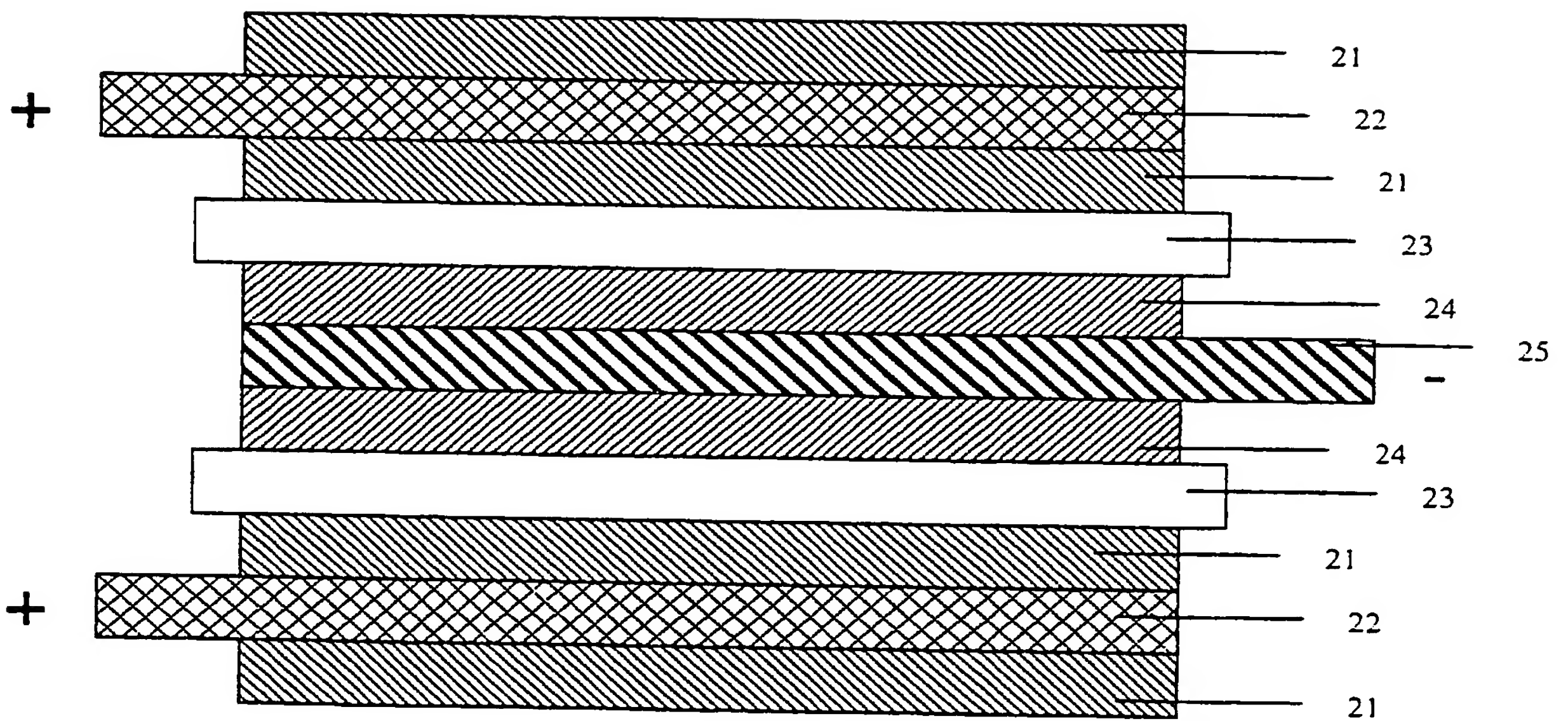
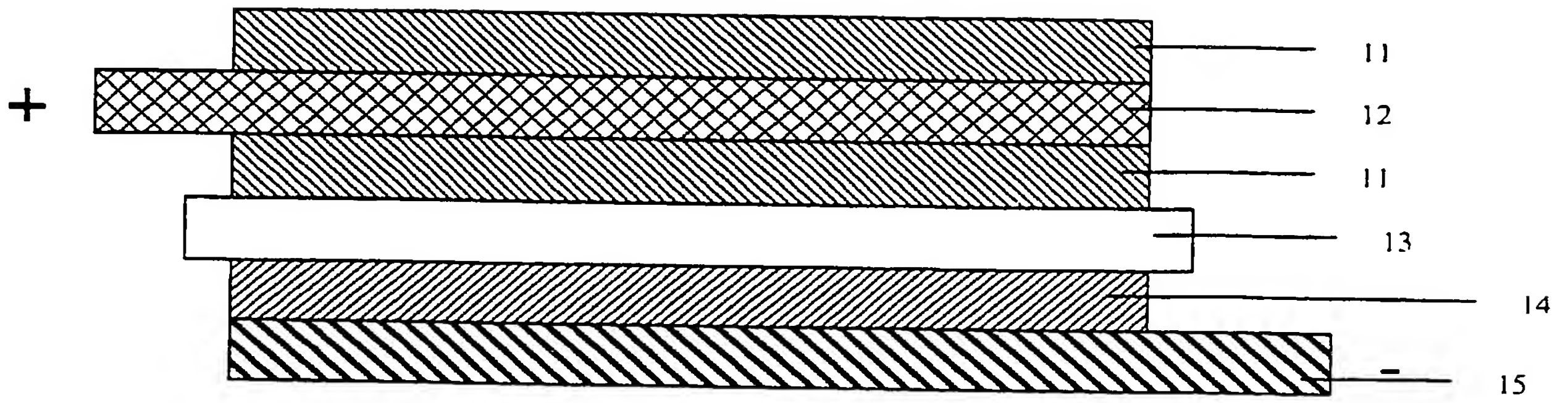
placing said device in the field of view of a thermographic camera;

applying a predetermined current and voltage to said conductive layers of said device;

observing through said thermographic camera any increases in surface temperature in said device which indicate the location of a short circuit.

15. A method according to claim 14, wherein said device is selected from the group consisting of primary batteries, rechargeable batteries, supercapacitors and electrochromic elements.

16. A method according to claim 14, wherein said device is a multi-layer bonded rechargeable plastic lithium ion battery.



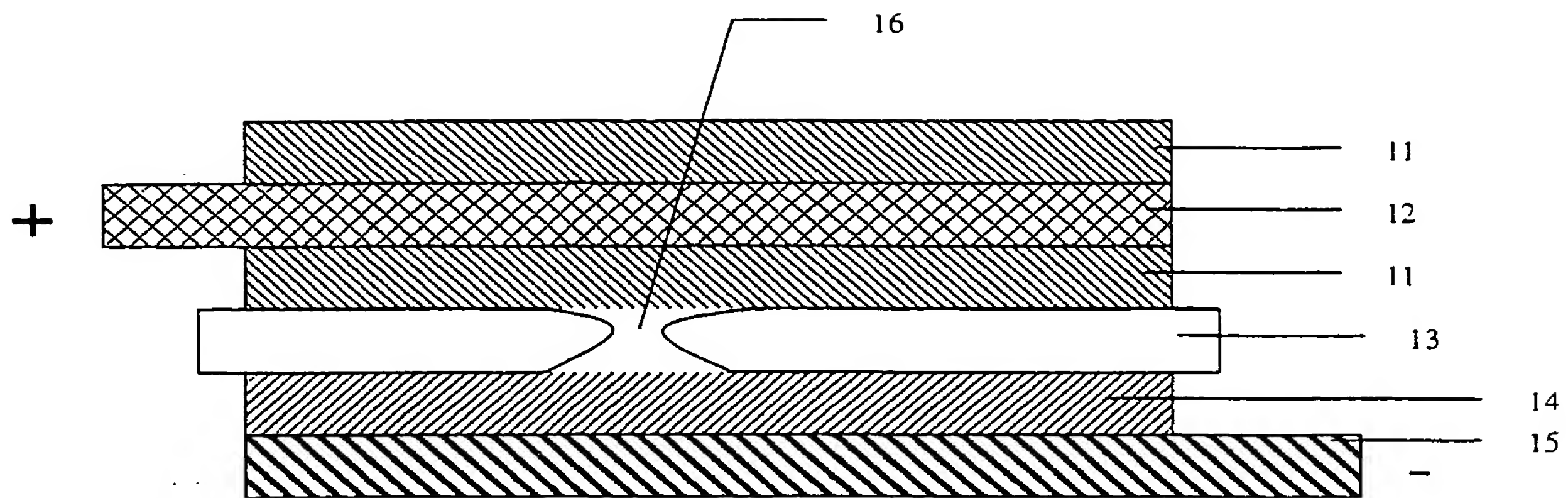


FIG. 2

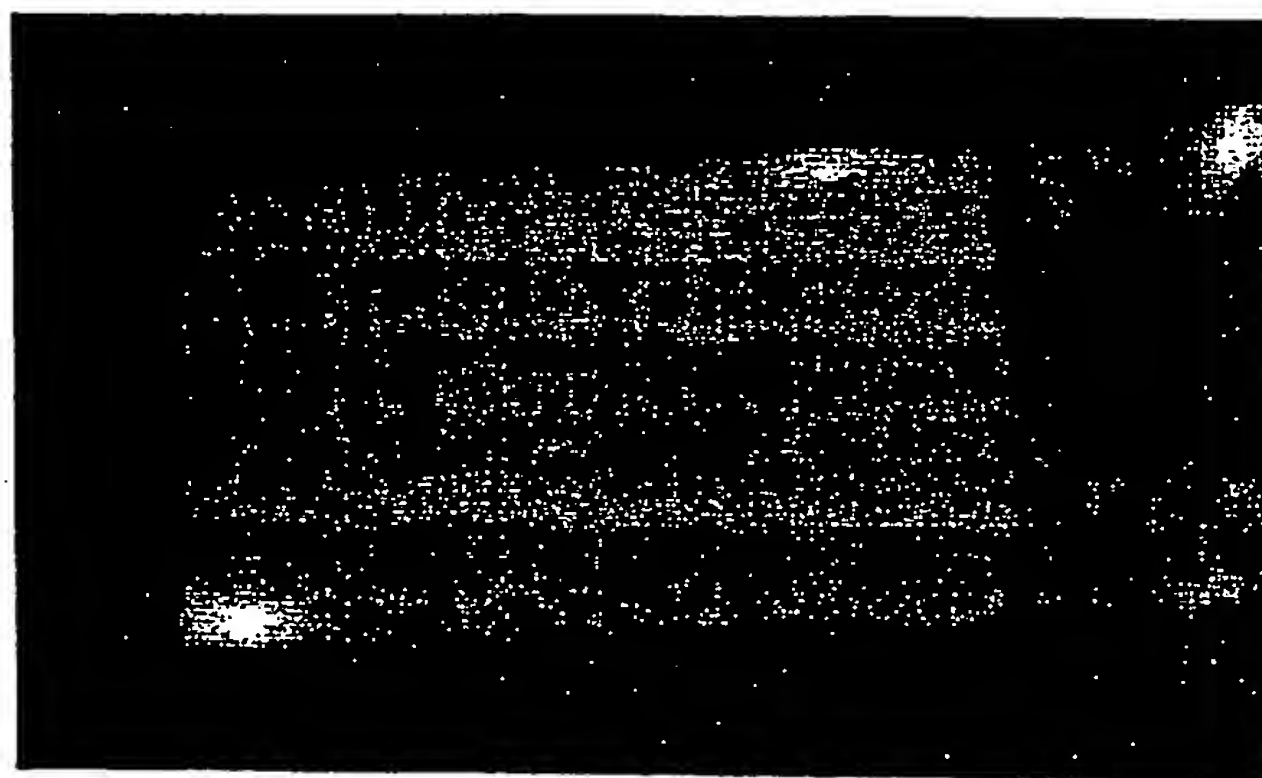


FIG. 3



## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US00/15635

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) : G 01 N 25/72

US CL : 374/4,5,6,7; 250/330,340,341.1; 324/501

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 374/4,5,6,7; 250/330,340,341.1; 324/501

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

EAST

search terms: infrared camera, current, defects, conductive layers, battery, separator layer, temperature, inductive heating

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|-----------|--|-----------------------|
| X         | US 5,483,068 A (Moulton et al.) 09 January 1996 (09.01.1996), col. 6, lines 13-44. | 1-16                  |
| A         | US 5,032,727 A (Cox, Jr. et al.) 16 July 1991 (16.07.1991), col. 3, lines 18-44    | 1, 14                 |
| A         | US 5,089,700 A (Sapia et al.) 18 February 1992 (18.02.1992), col. 2, lines 30-61   | 1, 14                 |
| Y         | US 4,431,967 A (Nishioka) 14 February 1984 (14.02.1984), col. 4, lines 55-62       | 1, 14                 |
| Y         | US 5,504,017 A (Yue et al.) 02 April 1996 (02.04.1996), col. 5, lines 12-32        | 1, 14                 |

☒ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

|   |  |
|---|--|
| * Special categories of cited documents:  | "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention  |
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| "P" document published prior to the international filing date but later than the priority date claimed  |  |

Date of the actual completion of the international search

14 JULY 2000

Date of mailing of the international search report

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## INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US00/15635

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|-----------|--|-----------------------|
| A         | JA 57-79463 A (Yamada) 18 May 1982 (18.05.1982), abstract                          | 1, 14                 |
| A         | JA 58-99767 A (Yabe) 14 June 1983 (14.06.1983), abstract                           | 1, 14                 |